

Estimate of nitrogen oxide emissions from shipping by satellite remote sensing

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[1] In recent years, the role of ship emissions has attracted increasing attention. Recent inventories estimate that they contribute significantly to anthropogenic emissions of nitrogen oxides, with a large impact on composition and chemistry of the marine boundary layer. Nevertheless, the number of observational studies is still rather small. Here we present the first detection of ship tracks in NO₂ maps derived from satellite data. We have used the congested track connecting Sri Lanka to Indonesia to estimate the corresponding ship emissions of nitrogen oxides to 23 (10–73) Gg [N]/yr. In addition, we were able to derive the mean lifetime of boundary layer NO_x to be 3.7 (1.9–6.0) hours in the respective region. Our estimates are in good agreement with ship emission inventories (about 22–54 Gg [N]/yr) as well as the short lifetimes (of down to 2 hours at daytime) as recently modeled for ship plumes.

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1. Introduction

[2] Nitrogen oxides (NO + NO₂ = NO_x) are important trace gases in the troposphere, driving catalytic ozone production and the oxidising power of the atmosphere. Global production is estimated to be on the order of 23–81 Tg [N]/yr, approx. half of it due to fossil fuel combustion [Lee *et al.*, 1997]. Recent inventories estimate ship emissions to contribute 3.6 [Endresen *et al.*, 2003] to 6.9 Tg [N]/yr [Corbett and Köhler, 2003]. These numbers reflect their importance as well as the still high uncertainty. Model calculations postulate a remarkable change in tropospheric composition due to ship emissions [e.g., Lawrence and Crutzen, 1999; Davis *et al.*, 2001; Endresen *et al.*, 2003; von Glasow *et al.*, 2003], since they are the only anthropogenic source of NO_x in the marine boundary layer, and the effect of additional NO_x sources on ozone production is larger for low NO_x background levels. Recent studies analysed the dispersion and chemical evolution of ship plumes [Davis *et al.*, 2001; Song *et al.*, 2003; von Glasow

et al., 2003] and point out the importance of in-plume-chemistry: as a result of ship emissions, daytime levels of OH as well as nighttime levels of NO₃/N₂O₅ are increased drastically, thus reducing the lifetime of NO_x. Model calculations estimate in-plume lifetimes to be about 2–6 hours (daytime) and 10–20 hours (nighttime), respectively [Song *et al.*, 2003; von Glasow *et al.*, 2003]. As a consequence, global models tend to overestimate NO_x concentrations in and close to ship tracks [Kasibhatla *et al.*, 2000] as they do not resolve in-plume processes. However, the number of observational studies for evaluating ship inventories as well as their impact on chemical composition and processes is still rather small.

[3] Satellite based measurements of atmospheric trace gases are a powerful enrichment to measurements from conventional platforms. They allow global monitoring with uniform instrumental features in long time series as well as the identification and quantification of different NO_x sources like biomass burning [Spichtinger *et al.*, 2001; Leue *et al.*, 2001], lightning [Richter and Burrows, 2002; Beirle *et al.*, 2004a] or anthropogenic emissions [Martin *et al.*, 2003; Beirle *et al.*, 2003, 2004b]. For remote ocean areas, expected pollution levels due to ship emissions are rather low compared to industrialized areas and thus hard to detect. Thus we make use of the specific advantages of satellite observations, namely their good spatial and temporal coverage, allowing the identification of characteristic patterns, even if faint, in the retrieved maps.

2. GOME Retrieval

[4] For this study we have used data from the Global Ozone Monitoring Experiment (GOME) [Burrows *et al.*, 1999]. GOME orbits the earth onboard the ERS-2 satellite, flying in a sun-synchronous nearly polar orbit and crossing the equator at 10:30 a.m. local time. The GOME instrument consists of four spectrometers measuring the radiation reflected by the earth in the UV/vis spectral range (240–790 nm) with a resolution of 0.2–0.4 nm. The extent of a GOME ground pixel is 320 km east-west and 40 km north-south (size and orientation of a GOME pixel are illustrated in Figure 1b). Within three days, global coverage is achieved at the equator.

[5] The GOME raw spectra are analysed at 430–450 nm to derive slant column densities of NO₂ [Wagner, 1999] by applying the established Differential Optical Absorption Spectroscopy (DOAS) [Platt, 1994]. The retrieval of tropospheric column densities from total columns is described in several studies [e.g., Leue *et al.*, 2001; Wenig, 2001; Martin *et al.*, 2002]. For the present study we have estimated the stratospheric fraction over the remote Pacific [Richter and

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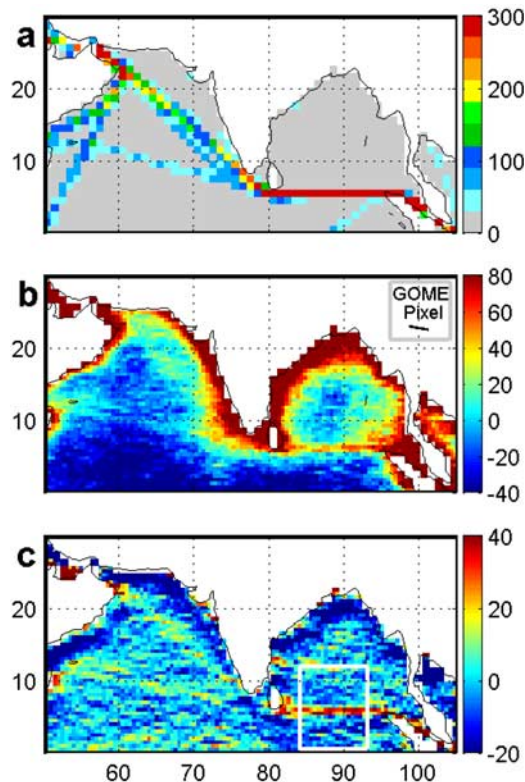


Figure 1. NO_x emissions from ships in the Indian ocean. (Land masses are masked out.) a) Inventory data (mg [N] per m² and year) by Endresen *et al.* [2003], using the AMVER (Automated Mutual-assistance Vessel Rescue system) distribution of ship reporting frequencies, weighted by the ship size (O. Endresen, personal communication, 2004). b) Tropospheric vertical column densities of NO₂ (10¹³ molec/cm²). GOME measurements of spring 1996–2001 with cloud fractions below 5% are averaged. The narrow ship track between Sri Lanka and Indonesia is clearly visible. For illustration, size and orientation of a single GOME pixel is displayed. c) High-pass filtered (along latitudinal component) NO₂ VCD. Additionally, the two major tracks west of India emerge. The box (84°–93°E, 0°–12°N) indicates the data used for the quantitative estimate of NO_x emissions from ships (see text).

Burrows, 2002] and subtracted it to yield tropospheric columns. To retrieve tropospheric vertical column densities (VCDs), radiative transfer modeling (RTM) has to be applied. We use the Monte-Carlo-RTM TRACY [von Friedeburg, 2003] with the following boundary conditions: a sea surface albedo of 5% (at 440 nm) [Koelemeijer *et al.*, 2003], a characteristic sea salt aerosol profile, and NO₂ well mixed in the maritime boundary layer (MBL) with a typical height of 700 m (500 m–1000 m). The overall uncertainty of the RTM correction is about 25%, to which the influence of albedo, aerosols and MBL contribute approx. 5%, 10%, and 10%, respectively. In this study, we only consider cloud free pixels, i.e., the cloud fraction (taken from the Heidelberg Iterative Cloud Retrieval Utilities HICRU [Grzegorski, 2003]) being less than 5%. The remaining cloud effect (mostly shielding) on the tropospheric VCD is less than 12%. For the considered 6 years of GOME data

(1996–2001), on average about 180 observations (out of 700) remain for each pixel.

3. NO_x Emissions From Ships

[6] According to recent studies on ocean going ship traffic [Corbett *et al.*, 1999; Endresen *et al.*, 2003], the Atlantic ocean is the most travelled ocean of the world, but ship tracks are distributed quite homogenously. In contrast, in the Indian ocean between Sri Lanka and Sumatra, all ships cruise a single uniquely narrow track (Figure 1a). Due to this feature, this particular track is clearly detectable in the tropospheric GOME data (Figure 1b). In Figure 1c we additionally show the high-pass filtered signal (recursive filter with a cut off frequency of 1/(1000 km), applied to the latitude coordinate), also indicating the ship routes connecting India and the Gulf of Aden and the Gulf of Oman, respectively. The nearly perfect agreement between the trace gas patterns and the ship routes allows the doubtless identification of ship emissions as source.

[7] For a quantitative analysis, we have considered the box indicated in Figure 1c and calculated zonal means (Figure 2). For all seasons, the sections show a clear maximum at 6°N. However, the shape of the peak differs significantly for the seasons and shows a strong asymmetry for summer (i.e., June–August) and winter (December–February). This is due to the fact that the ship track is traversed by the Intertropical Convergence Zone (ITCZ) twice a year. As a result, the mean wind directions of summer and winter are nearly opposite. The mean meridional wind component is 6.5 m/s in summer and −4 m/s in winter (NCEP/NCAR reanalysis [Kanamitsu *et al.*, 2002]). The respective curves are broadened to the North (summer) and the South (winter) respectively, while

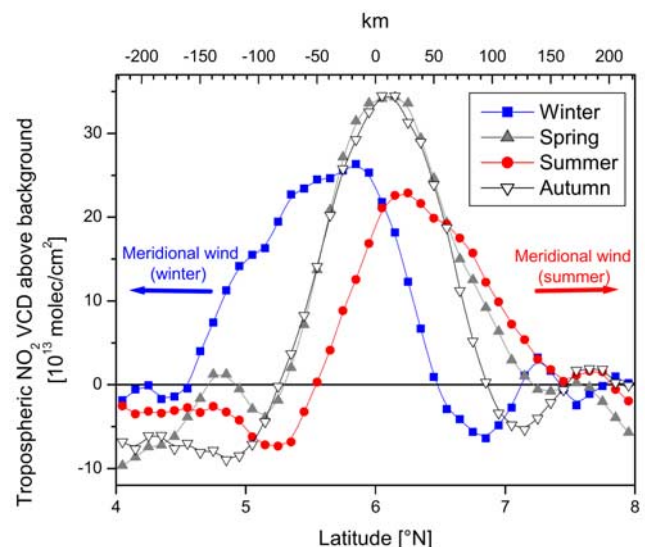


Figure 2. High-pass filtered tropospheric NO₂ VCD, zonally averaged over the box in Figure 1c, as function of latitude (below) with an additional km-scale (above). For all seasons, a clear maximum at the ship track position (≈6°N) can be seen. The graphs for winter and summer are asymmetric due to strong meridional winds, indicated by the arrows.

they are quite symmetric in spring and autumn when meridional wind speeds are lower (1–2 m/s). The decay of the downwind NO₂ VCD reflects the photochemical degradation, thus the actual peak shape allows us to deduce the mean lifetime of boundary layer NO_x. We used a least squares method to fit a Gaussian distribution (reflecting possible scatter of the source, diffusion of the pollutants, and convolution with 40 km GOME extent), convoluted with an exponential distribution, accounting for the first order loss of NO₂. The resulting 1/e-lengths are 90 km for summer and 122 km for winter with an uncertainty of 20%. The corresponding lifetimes τ (obtained by dividing by the respective wind speeds) are 3.8 hours in summer and 8.5 hours in winter. This large difference cannot be explained merely by variations of temperature, ozone concentrations or photolysis rates. The shorter summer lifetime is probably a result of the different meteorological conditions, i.e., higher water vapour concentrations (as can be seen e.g., from GOME) as well as higher precipitation rates (NCEP/NCAR reanalysis) in summer. Therefore we expect higher OH concentrations as well as maybe increased scavenging of N₂O₅, thus implying a shorter lifetime. However, assuming the ship emissions to be constant throughout the year, the ratio of the lifetimes for winter and summer should be directly reflected in the ratio of the respective integrals over the sections in Figure 2. The latter is 1.33 (whereas spring and autumn give the same integral as winter), indicating that the lifetime is indeed larger in winter, but rather by 33% than by 100%. We therefore expect the summer and winter lifetimes to be comparable, and take the retrieved lifetimes for summer and winter as representative for a lower and upper bound of the overall mean lifetime, and their average (6.15 hours) as best guess.

[8] Of course fitting a mean lifetime for a whole season is a simplification, since the instantaneous lifetime varies strongly over time. The largest effect, however, is the diurnal variation between noon (about 2–6 hours) and midnight (10–20 hours) and nearly infinite values during dawn as modeled by Song *et al.* [2003] and von Glasow *et al.* [2003]. The “equivalent” mean lifetime (defined as the lifetime giving the correct integral over the decay curve) is dominated by the low daytime value (for instance, assuming τ to be 2 hours during the day, 10 hours at night, and infinite during dawn results in a “equivalent” mean lifetime of 3.4 hours). However, since the GOME measurement is at 10:30 local time, it is significantly influenced by the accumulated pollution during dawn, resulting in quite high lifetime estimations: We calculated spatial NO₂ distributions (assuming a constant source at 6°N and a constant wind speed of 5 m/s) based on diurnal cycles of the instantaneous lifetime from Song *et al.* [2003] and from the model runs by von Glasow *et al.* [2003], and then fitted a mean lifetime in the same way as to the GOME data. In these calculations, we generally overestimate the “equivalent” mean lifetime by 43%–100%. We therefore have to downscale our estimated lifetime by 0.6 (0.5–0.7), resulting in $\tau = 3.7$ (1.9–6.0) hours.

[9] We use this mean lifetime to quantify absolute NO_x emissions from ships. Integrating the annual mean signal from 4.5°N to 7.5°N and multiplying with the box width of 9°, i.e., 995 km (at 6°N), we find the daily enhancement to

be $3.0 \cdot 10^{29}$ molecules [NO₂]. To deduce daily emissions from the observed VCD, we have to scale this number with 24 h/ τ [hours], resulting in a factor of 6.5. Furthermore, the ratio of NO₂ to total NO_x at 10:30 local time is about 0.72 (0.67–0.77) (modeled by von Glasow *et al.* [2003]), giving a further factor of 1.39. The resulting ship emissions in the respective box are $2.7 \cdot 10^{30}$ molec [NO_x]/day or 63 t [N]/day, i.e., 23 Gg [N]/year.

[10] Due to the high pass filtering, errors arising from the estimate of stratospheric NO₂, superimposed NO_x (lightning and/or transport of continental pollution) and the degradation of the GOME instrument (A. Richter and T. Wagner, Diffuser plate spectral structures and their influence on GOME slant columns, available at <http://www-iup.physik.uni-bremen.de/gome/data/>, 2001) are nullified. Thus, the remaining uncertainties arise from the RTM (~25%), the precise recognition of the ship induced enhancement in the raw data (~25%), the NO₂/NO_x ratio (~8%), and the lifetime estimate (~50%). Thus we derive the emissions to be 10–73 Gg [N]/year by taking the minimum/maximum values for all factors. The largest contribution to the overall uncertainty arises from the lifetime estimate. However, additional information on this quantity would significantly reduce our uncertainty to about 16–37 Gg [N]/year (assuming $\tau = 3.7$ hours as correct value).

[11] In principle, the GOME observations underestimate the actual VCD in the first stage of plume evolution (when the NO_x is close to the ground) due to an inappropriate RTM correction factor. However, the expansion of the plume is quite fast: after 6 minutes the plume height is already ~200 m (see equation 2 of von Glasow *et al.* [2003]). The fraction of NO₂ that we miss is below 5%, thus negligible.

[12] Table 1 compares our estimate to recent emission inventory datasets. Since the inventory of Corbett and Köhler [2003] is not spatially resolved, we derive a simple estimate by scaling the prior (spatially resolved) numbers of Corbett *et al.* [1999] with the ratio of 5.00 Tg N (global cargo fleet emissions as estimated by Corbett and Köhler [2003]) and 3.08 Tg N (total global emissions [Corbett *et al.*, 1999]), yielding a factor of 1.62. Overall, we find a good agreement of our results with the literature values. Please note that these numbers are affected by the underlying total global emission estimate as well as the assumed ship traffic distribution that may be biased for specific routes (see the discussions of Corbett and Köhler [2003] and Endresen *et al.* [2003]).

[13] Apart from the three tracks indicated in Figure 1c, we could not detect any other ship track in the GOME data, mainly for two reasons: a) the Indian ocean tracks are extremely narrow (in contrast to the smeared out Atlantic routes) and b) they have the peculiarity of being oriented in the east-west direction, almost parallel to the GOME pixel. Thus, the large east-west extent of the GOME pixel has no unfavourable effect.

4. Conclusions

[14] In this study we present the first detection and quantification of ship NO_x emissions from satellite. The line of enhanced NO₂ in the Indian ocean as seen in the 6-year composite of cloud free GOME observations clearly

Table 1. Total NO_x Emissions for the Indian-Indonesian Ship Track (84°–93°E, 0°–12°N)

Source	NO _x Emissions (Gg [N]/year)
EDGAR 3.2	34
[Olivier and Berdowski, 2001]	
Corbett et al. [1999] ^a	22
Corbett and Köhler [2003]	36 ^b
Endresen et al. [2003] ^c	41 ^d
	54 ^e
This study	23 (10–73) ^f
	23 (16–37) ^g

^aData kindly provided by J. Corbett, see <http://www.ocean.udel.edu/cms/jcorbett/sea/nitrogen.html>.

^bEstimated by simply scaling up the overall ship emissions of NO_x by Corbett et al. [1999] by a factor of 1.62 (see text).

^cData kindly provided by O. Endresen.

^dUsing the ship distribution from the Automated Mutual-assistance Vessel Rescue system (AMVER).

^eAMVER distribution weighted by ship size.

^fIncluding lifetime uncertainties.

^gAssuming 3.7 hours as correct mean lifetime.

coincides with the distinct ship track from Sri Lanka to Indonesia. We used seasonal differences in the wind direction to derive the mean lifetime of ship exhaust NO_x to be about 3.7 (1.9–6.0) hours. Our result (arrived at using an independent method) confirms recent modeling studies that predict rather low NO_x lifetimes within ship plumes. Using our lifetime estimate, we derived the total emissions within the considered area to be 23 (10–73) Gg [N]/yr in good agreement with recent inventories.

[15] Consequently, our new and independent method helps verify existing inventories as well as model calculations. Prospective refinements of our method are likely to reduce current uncertainties. Furthermore, successors of GOME like the **SC**anning **IM**aging **AB**sorption **SP**ectro**M**eter for **AT**mospheric **CH**artograph**Y** (SCIAMACHY), the **O**zone **M**onitoring **I**nstrument (OMI) and GOME II with improved spatial resolution might even allow the detection of further ship tracks (A. Richter et al., Satellite measurements of NO₂ from international shipping emissions, submitted to *Geophysical Research Letters*, 2004).

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